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Supplementary Material:

for “Structure-property relations of binary ferrite melts”

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1. EPSR model set up.

43BaO-57FeO_x at 1873K							
Gas	X	Ba (%) #atoms	Fe^{2+} (%) #atoms	Fe^{3+} (%) #atoms	O (%) #atoms	Total #atoms	Density ($Å^{-3}$)
O2	1.47	(21.5) 1290	(1.6) 96	(21.6) 1296	(55.3) 3318	6000	0.052
0.1% O_2	1.40	(21.5) 1290	(4.6) 276	(19.3) 1158	(54.6) 3276	6000	0.052
CO_2	1.37	(21.5) 1290	(6.5) 390	(17.8) 1068	(54.2) 3252	6000	0.052
$COCO_2$	1.18	(21.5) 1290	(17.0) 1020	(9.5) 570	(52.0) 3120	6000	0.052
99CO	1.05	(21.5) 1290	(25.1) 1506	(2.9) 174	(50.6) 3030	6000	0.053

43SrO-57FeO_x at 1873K							
Gas	X	Sr (%) #atoms	Fe^{2+} (%) #atoms	Fe^{3+} (%) #atoms	O (%) #atoms	Total #atoms	Density ($Å^{-3}$)
O2	1.48	(21.5) 1290	(0.1) 6	(22.7) 1362	(55.7) 3342	6000	0.0620
0.1% O_2	1.43	(21.5) 1290	(0.3) 18	(22.6) 1356	(55.6) 3336	6000	0.0620
CO_2	1.40	(21.5) 1290	(0.4) 24	(22.5) 1350	(55.6) 3336	6000	0.0620
$COCO_2$	1.23	(21.5) 1290	(1.9) 114	(21.4) 1284	(55.2) 3312	6000	0.0620
99CO	1.07	(21.5) 1290	(7.3) 438	(17.2) 1032	(54.0) 3240	6000	0.0620

Table T1. EPSR model compositions.

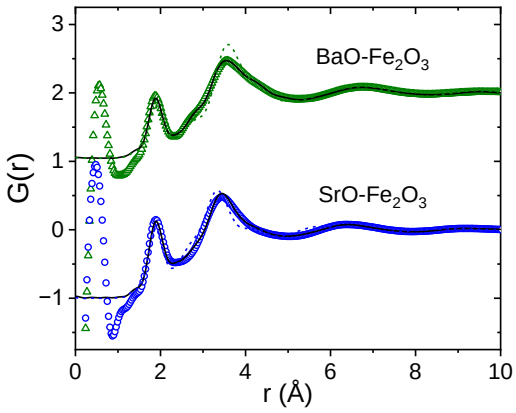

Figure S1. X-ray pair distribution functions of samples measured in oxygen at 1873 K (symbols). Compared to the EPSR models obtained using only the starting Lennard Jones potentials and effective Coulomb charges given in table II (dashed lines), and the improved EPSR models with the empirical potential turned on (solid lines).

2. EPSR model fits to diffraction data.

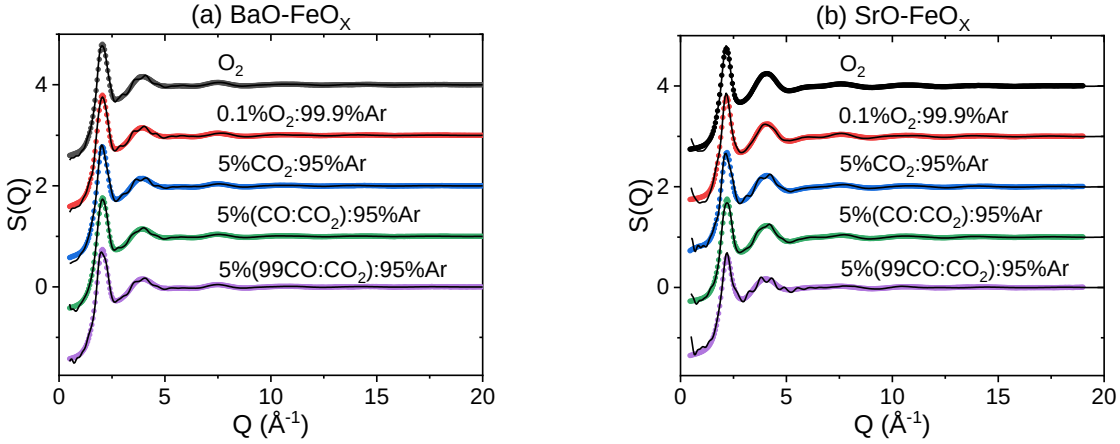

Figure S2. High energy x-ray structure factor (circles) and EPSR model fits (lines) to the (a) BaO-FeO_X and (b) SrO-FeO_X melts.

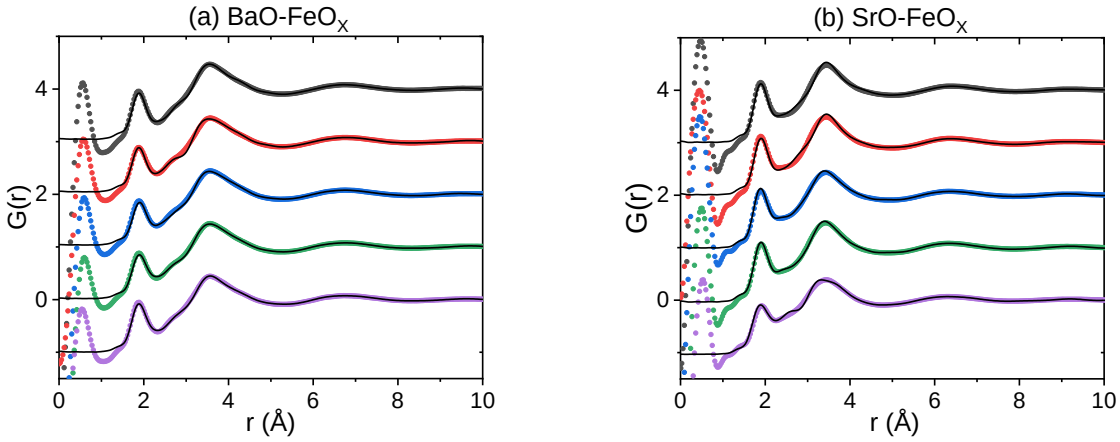

Figure S3. X-ray pair distribution function (circles) and EPSR model fits (lines) to the (a) BaO-FeO_X and (b) SrO-FeO_X melts.

3. Fe^{3+}/Fe^{2+} ratio from the activity scaling model applied to CaO-FeO_X melts.

The validity of this result can be checked by comparing the *Fe_xO* activity in CaO compared to BaO melts. However, here the activity of *Fe_xO* for *CaO-Fe_xO* melts reported from an earlier paper by Iwase *et al.* [1] are in slight disagreement with those values reported by Banya *et al.* [2], Chavez *et al*. [3] and earlier studies (~10% higher at X_{FexO}=*0.57*). Therefore, here we use the *FexO* activity data of Banya *et al*. [2] to perform the linear scaling factor approximation of CaO compared to BaO iron oxide melts which yields $a_{FexO}(BaO) \approx a_{FexO}(CaO)$. Combining equations (3), (4) and (6) for X_{FexO}=*0.57* we find that,

$$Fe^{3+}/Fe^{2+}(CaO) = 0.36\ Fe^{3+}/Fe^{2+}(BaO) \qquad (8)$$

Our previous measurements at the *43CaO-57FeO_X* composition (Shi *et al*. [4]) calculated the Fe^{3+}/Fe^{2+} ratio using equation 12 reported by Jayasuriya *et al*. [5], based on fitting Mossbauer data in Fe silicate glasses, see figure 3. These results are compared to those obtained using equation (8) and shown in figure S3.

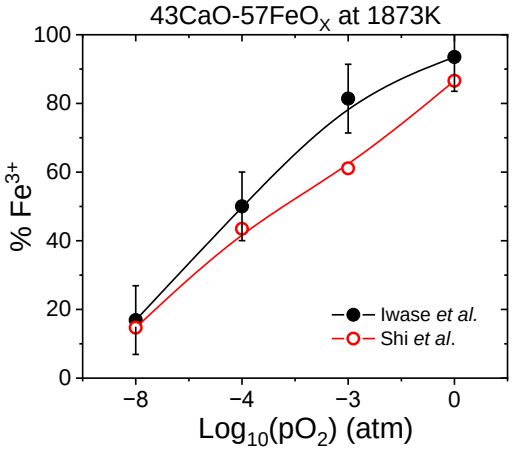

Figure S4. The *%Fe*$^{3+}$ as a function of oxygen partial pressure in *43CaO-57FeO*$_X$ melts obtained using equation (8) (black closed circles, Iwase *et al*. [6] using *Fe*$_x$*O* activity data from Banya *et al*. [2]) compared to Jayasuriya *et al*. [5] equation (12) (red open circles). The gas compositions are given in table 1. Based on the variation in *Fe*$_x$*O* activity we estimate an error of ±10% in %Fe^{3+}.

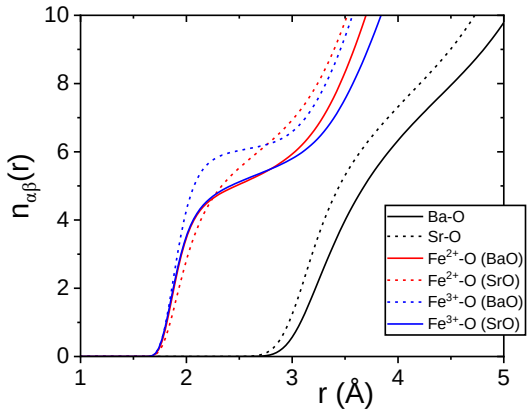

Figure S5. Running coordination numbers for 43MO-57Fe_2O_3 at 1873K where the M=Ba melt is shown as solid lines and the M=Sr melt is shown as dashed lines when exposed to the intermediate 5%(CO:CO_2):95%Ar gas environment.

References.

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